The Interaction of Water and Aerosols in the Marine Boundary Layer: A Study of Selected Processes Impacting Radiative Transfer and Cloudiness

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LONG-TERM GOALS

The overarching, log term goal of the study is to explore the profound effect of aerosol-water interaction both on radiation propagation in, and the thermodynamic structure of, the marine boundary layer. Specific goals are: 1) compile a climatology of aerosol hygroscopicity for use in the NAAPS and COAMPS models, and, further, to develop a model parameterization of hygroscopicity based on aerosol size and composition for such models, 2) explore the relative impacts of cross-inversion mixing and sub-cloud aerosol on cloud thickness and cloud base height, 3) quantify and parameterize the impact of precipitation scavenging on below cloud radiative transfer and cloud liquid water path. The sampling platform utilized is the CIRPAS Twin Otter research aircraft and the venue is the littoral environment off the California coast, representative of areas with high shipping densities.

OBJECTIVES

During the current year, we have concentrated primarily on finishing up the analysis of data gathered during the CARMA-III campaign and on the preparation which was necessary for the just completed CARMA-IV field program. The former goal has been essentially completed, culminating in a publication (listed below). The latter objective constitutes the real commencement of the current grant and we concentrate on it in the following discussion.

The goals of the CARMA IV field campaign, in addition to the obvious one of ensuring that the instrumentation package for the Twin Otter aircraft was working properly, are as follows:

- Assemble a database on the size-resolved aerosol hygroscopicity for marine air off of the California coast

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14. ABSTRACT

The overarching, log term goal of the study is to explore the profound effect of aerosol-water interaction both on radiation propagation in, and the thermodynamic structure of, the marine boundary layer. Specific goals are: 1) compile a climatology of aerosol hygroscopicity for use in the NAAPS and COAMPS models, and, further, to develop a model parameterization of hygroscopicity based on aerosol size and composition for such models, 2) explore the relative impacts of cross-inversion mixing and sub-cloud aerosol on cloud thickness and cloud base height, 3) quantify and parameterize the impact of precipitation scavenging on below cloud radiative transfer and cloud liquid water path. The sampling platform utilized is the CIRPAS Twin Otter research aircraft and the venue is the littoral environment off the California coast, representative of areas with high shipping densities.

15. SUBJECT TERMS

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- Assess the relative importance of mixing of super-inversion air, and below cloud aerosol concentration on the cloud optical thickness and albedo. One particularly interesting facet of this assessment would be an appraisal of the impact of inhomogeneous mixing on the CDNC and cloud albedo (cf., Burnet and Brenguier, 2006). From this, assess the possibility of parameterizations that would predict the impact of these processes on the LWP and cloud base height in the MBL

- Assess the importance of elemental carbon (EC) particles as CCN

- Evaluate the impact of chemistry on aerosol hygroscopicity during vertical transport in the MBL

APPROACH

Obtaining reliable measurements of both super-micron aerosol particles and their hygroscopicity is a major goal of our program. Our primary tool for this goal is the AHS described in last years annual report, coupled with the Descriptive Hygroscopic Growth Factor (DHGF) analysis technique also described in that report. For the association of the measured hygroscopicity as a function of size with chemical composition, we have used several different substrate-based approaches for measuring the aerosol chemistry.

Assessing the relative importance of mixing of super-inversion air, and below cloud aerosol concentration on the cloud optical thickness and albedo is our second objective. One particularly interesting facet of this assessment would be an appraisal of the impact of inhomogeneous mixing on the CDNC and cloud albedo using the approach of Burnet and Brenguier (2006). From this, assess the possibility of parameterizations that would predict the impact of these processes on the LWP and cloud base height in the MBL.

The methodology for achieving the third objective involves the use of two instrument new to our program. The first of these is the annular geometry CCN spectrometer manufactured by DMT Inc. based on the design of Roberts and Nenes (2005). This will yield a continuous record of the CCN concentration at five supersaturations with a time resolution of about 10 minutes. This data will be compared with the concentration of EC bearing particles measured by the SP2 instrument manufactured by DMT Inc. and recently evaluated and described by Moteki and Kondo (2007). The comparison will yield the fraction of the CCN number concentration at each supersaturation that contain EC.

The final objective, evolution of the aerosol hygroscopicity in the vertical due to chemical activity will be examined by comparing both the hygroscopicity and chemistry of the aerosol at different altitudes in the MBL as per Hegg et al (2007).

WORK COMPLETED

The CARMA-IV field project, the main data source for the analyses proposed above, took place during August of this year. Fourteen airborne missions were successfully undertaken, resulting in a substantial database for the analyses proposed above. It should be noted, however, that one tentative objective of our grant (though not listed above), the evaluation of the impact of precipitation scavenging on the MBL aerosol concentration, could not be addressed with the data available and has been dropped from further consideration. Essentially, the highly atypical weather encountered during the field program does not permit an assessment of this important issue.

Currently, we are evaluating our data and doing chemical analysis on the various chemical filter/substrate samples obtained in the field. While it is thus premature to undertake detailed analyses, a few preliminary results are already available and we present them briefly below.

RESULTS

Pursuant to our first goal, DHGF spectra were obtain for a number of aerosols with presumably different hygroscopicities. Three examples of this diversity are shown in Figure 1.

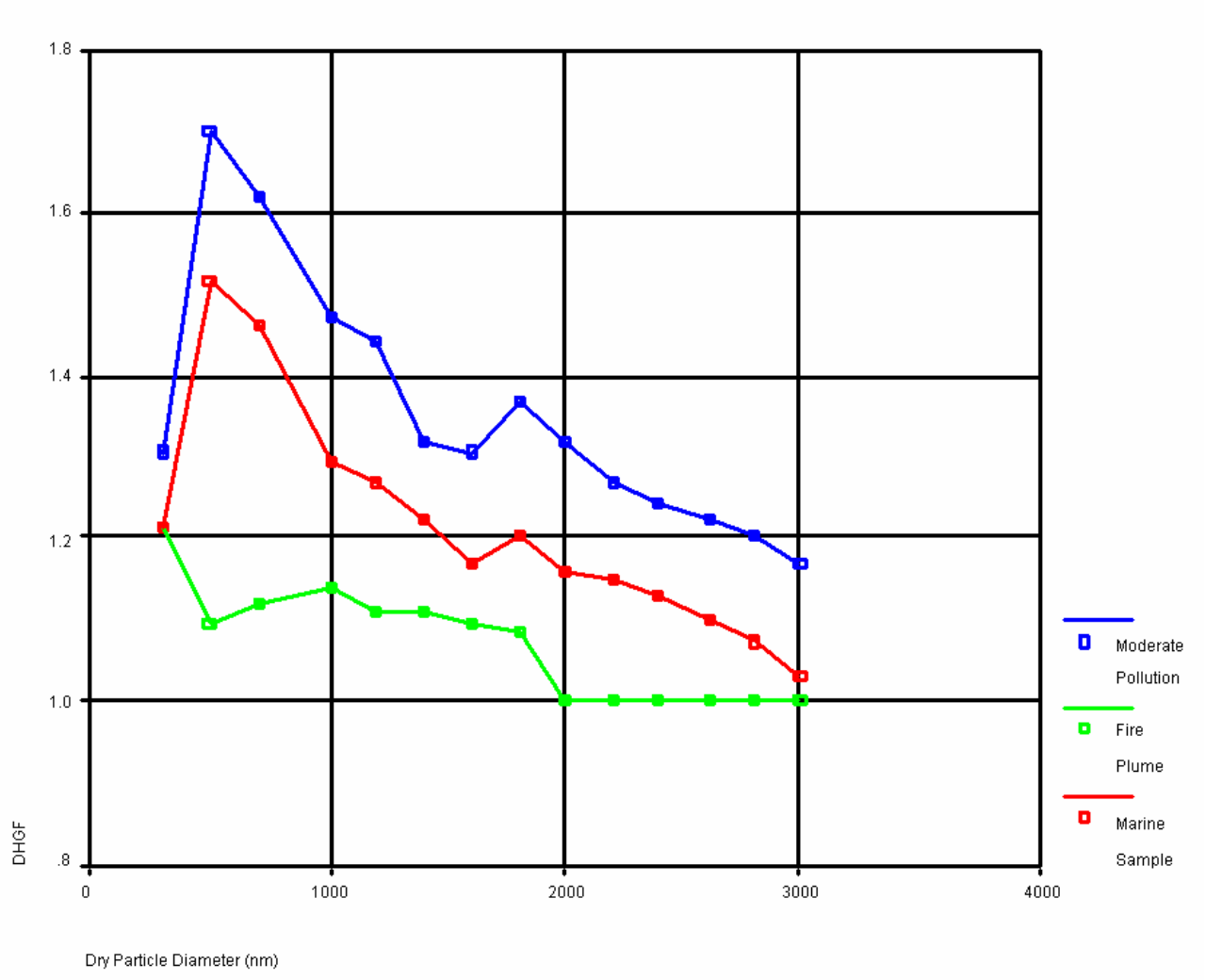

Figure 1. DHGF spectra measured during CARMA-IV for three different aerosol types. All the spectra show relatively low hygroscopicity at larger sizes but differ widely for submicron sizes, with the fire haze sample showing the lowest hygroscopicity.

The relatively clean marine spectrum shown is quite similar to those obtained during CARMA-III and reported in Hegg et al (2006). Also shown in the figure is a spectrum for a sample taken in the regional haze associated with the very large fire near Santa Barbara which impinged on our operational area. This spectrum shows the haze aerosol to have substantially reduced hygroscopicity, as expected, and a

flatter overall spectrum suggesting a disproportionate reduction in the hygroscopicity of the sub-micron particles, as seen in an earlier fire plume sample (Hegg et al, 2006). The DHGF spectrum for the moderately polluted aerosol sample also sows reduced hygropscopicity but is intermediate between the clean marine and fire haze spectra. While the chemical analysis of the aerosols for these spectra is not yet complete, it does appear that both the fire haze and pollution samples have a high proportion of insoluble mass, presumably organic.

In addition to the hygroscopicity measurements by the AHS, we continue to make and utilize measurements by our older instrumentation package, applying the data to the issues outlined above. For example, As in past CARMA experiments, a set of optical properties including the extensive properties, particulate scattering and absorption coefficients and scattering coefficient as a function of relative humidity, was measured continuously during the flights. Intensive properties, e.g., Ångström exponents of scattering and absorption, hygroscopic growth factor (g) and single scattering albedo (SSA), which are relatively independent of aerosol concentration and more closely related to the size distribution and chemical composition of the particles, were calculated from the measured parameters. The haze aerosol from the Santa Barbara forest fire, as suggested by the DHGF spectra, provided a contrast to the usual marine aerosol and occasional anthropogenic pollution plumes encountered in previous CARMA flights.

The intensive optical properties of the aerosol in the plume of the forest fire near the source (in terms of distance and time) and also the more aged aerosol that was entrained into the coastal MBL were markedly different from those of pollution plumes and marine aerosol. Scattering Ångström coefficient was relatively high, values of 2 to 3, similar to the values found in urban plumes but much larger than those of marine aerosol, ca. 0 to 0.5. Absorption Ångström was also high, values of 2 to 4, compared to values of 1 to 1.5 more commonly found in pollution plumes. (Absorption Ångström of marine aerosol is generally undefined because the absorption coefficients are too low to be measured with enough accuracy to permit the derivative with wavelength to me determined.) The high values are due to light absorbing organic compounds in the particles that absorb preferentially at the shorter wavelengths. Particles containing elemental carbon or soot in combination with other light absorbing carbon compounds dominate in aged urban pollution plumes and generally have a dependence that is inversely proportional to the first power of wavelength rather than higher powers. The single scattering albedo of the fire aerosol was low with values between 0.7 and 0.9 cf. marine or urban aerosols which have SSA of > 0.99 and 0.9 to 0.96, respectively. The scattering hygroscopic growth factor, g, was 0.1 or less compared to a growth factor of 0.5 or larger for marine and urban aerosols.

The combination of these parameters provides a “fingerprint” or tracer for the source and relative mix of aerosol present. The fingerprint is not as definitive as a set of chemical tracers but is rapidly and easily measured. The derived parameters for the Santa Barbara fire aerosol are consistent with a biomass burning aerosol particle size distribution that is relatively small and a chemical composition that is dominated by light absorbing organic compounds, high absorption Ångström low SSA, that are relatively insoluble (as suggested by the preliminary chemical analysis mentioned above), low hygroscopic growth factor. This state persisted for some time during atmospheric ageing even after the aerosol was entrained into the MBL and marine stratus.

IMPACT/APPLICATIONS

These preliminary results support our tentative conclusions from CARMA-III, namely, the hygroscopicity data as a function of aerosol size reveal that there are distinct patterns of DHGF for different aerosol type and illustrate the importance of chemical composition in the determination of

aerosol hygroscopicity. The high but rather systematic variability in hygroscopicity over size suggests that mean aerosol hygroscopicity my be a poor predictor of the impact of this hygroscopicity on other aerosol integral properties or of the impact of aerosols on other processes, such as gas/aerosol interactions.

TRANSITIONS

None.

RELATED PROJECTS

The size dependent hygroscopic growth of aerosols, including super-micron aerosols (cf., Quinn et al, 1998), is a major factor in both the radiative energy balance of the lower marine atmosphere and the propagation of radiation through the MBL. Such radiation properties are necessary parameters for numerical modelers developing prognostic models. It is, furthermore, an aerosol characteristic closely related to CCN activity and, indeed, such activity can be predicted from it. Hence, these measurements are highly relevant to determination of CCN spectra and thus of the microphysics of MBL clouds.

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PATENTS

None.